Magnetism and Superconductivity in (RE)Ni_2B_2C: The Case of $TmNi_2B_2C$

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Abstract

The recently reported[1,2] coexistence of an oscillatory magnetic order with the wave vector $Q = 0.241$ Å^{-1} and superconductivity in $TmNi_2B_2C$ is analyzed theoretically. It is shown that the oscillatory magnetic order and superconductivity interact predominantly via the exchange interaction between localized moments (LM's) and conduction electrons, while the electromagnetic interaction between them is negligible. In the coexistence phase of the clean $TmNi_2B_2C$ the quasiparticle spectrum should have a line of zeros at the Fermi surface, giving rise to the power law behavior of thermodynamic and transport properties. Two scenarios of the origin of the oscillatory magnetic order in $TmNi_2B_2C$ are analyzed: a) due to superconductivity and b) independently on superconductivity. Experiments in magnetic field are proposed in order to choose between them.

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The problem of the coexistence of magnetic order and superconductivity is a long-standing one and Ginzburg[3] was the first to note the antagonistic character of these phenomenon. Further impetus in this field came after the discovery of the ternary rare earth (RE) compounds (RE)Rh_4B_4 and (RE)Mo_6X_8 (X=S,Se), see Ref. 4. In many of these compounds both ferromagnetic (F) and antiferromagnetic (AF) ordering coexist with superconductivity (S), see Refs. 5,6. It turned out that the coexistence of S and AF ordering was realized in many of these compounds[7] usually up to $T = 0$, while S and modified F ordering coexisted in $ErRh_4B_4$, $HoMo_6S_8$ and $HoMo_6Se_8$ only. The reason for this was the antagonistic characters of these orderings. A theory has been developed and the phase diagram was given in Refs. 5,8,9 where the possibility of the coexistence of S and spiral or domain-like magnetic order has been elaborated quantitatively by including the exchange (EX) and electromagnetic (EM) interaction of conduction electrons and localized magnetic moments (LM's). It has been also demonstrated that the theory based on the EM interaction only[10] can not describe the coexistence problem in real systems. Note, some heavy fermions UPt_3,URu_2Si_2 etc. show a coexistence of the AF and S orderings. Recently, it has been also found experimentally the coexistence of nuclear magnetism and superconductivity[11] in $AuIn_2$, which was theoretically analyzed in Ref. 12.

However, recent discovery of superconductivity in the quaternary intermetallic compounds (RE)Ni_2B_2C (RE=Sc,Y,Lu,Tm,Er,Ho and Th) has received appreciable attention, because of the relatively high superconducting transition temperature - 16.6 K in Lu. Band structure calculations[13] show that the electronic spectrum is three-dimensional. Because of the spatial isolation of magnetic ions there is a possibility for the coexistence of a magnetic (M) order and S in (Ho,Er,Dy)Ni_2B_2C with $(T_c; T_M)$=(8,11,6.5 K;6,7,10.5 K) respectively[14]. T_M is the (antiferro)magnetic transition temperature. These compounds are characterized by the ferromagnetic alignment in each layer, with the magnetic moments of two consecutive layers aligned in opposite directions. Band structure calculations[15] of the nonmagnetic $LuNi_2B_2C$ compound show that the conduction electron density on the RE ions is small (similarly as in (RE)Rh_4B_4 and (RE)Mo_6X_8) giving rise to a relatively small exchange

energy.

The subject of this paper is the theoretical analysis of the coexistence problem in $TmNi_2B_2C$, which is superconducting below $T_c \approx 11$ K with an oscillatory magnetic ordering of the Tm moments below $T_M \approx 1.5$ K with persisting coexistence up to $T = 0$. The magnetic structure is incommensurate[1] with the Tm moments along the c-axis and with a sinusoidal modulation of their magnitudes along the (110) direction. This sinusoidal order is characterized by the wave vector $Q = 0.241$ Å^{-1}. One should stress the following facts: (1) $TmNi_2B_2C$ is unique in the (RE)Ni_2B_2C family, which shows a modulation of the magnetic order in the (110) direction and the alignment of the moments along the c-axis; (2) the wave vector Q is neither large nor small ($\xi_0^{-1}, \lambda^{-1} \ll Q \ll k_F$), where ξ_0, λ, k_F are the superconducting coherence length, magnetic penetration depth and Fermi momentum respectively.

In what follows the theory of magnetic superconductors - the MS theory[5,8], is applied to $TmNi_2B_2C$ and it will be shown that the competition between S and the oscillatory M order is *predominantly* due to the EX interaction, while the EM one is negligible, not only in this compound but also in the whole (RE)Ni_2B_2C family. An analysis of effects of the magnetic field allows us to discern between two possible scenarios for the origin of the oscillatory magnetic order.

The MS theory[5,8] considers all important interactions between $LM's$ and conduction electrons: (1) via the direct EX interaction; (2) via the induced magnetic field $\mathbf{B}(\mathbf{r}) = \mathrm{curl}\mathbf{A}(\mathbf{r})$ - the EM interaction, which is due to the dipolar magnetic field $\mathbf{B}_m(\mathbf{r}) = 4\pi\mathbf{M}(\mathbf{r})$. The general Hamiltonian of the (RE)Ni_2B_2C compounds is given by

$$\hat{H} = \int d^3r\{\psi^\dagger(\mathbf{r})\epsilon(\hat{\mathbf{p}}-\frac{e}{c}\mathbf{A})\psi(\mathbf{r}) + [\Delta(\mathbf{r})\psi^\dagger(\mathbf{r})i\sigma_y\psi^\dagger(\mathbf{r}) + c.c] + \frac{\mid\Delta(\mathbf{r})\mid^2}{V} + \hat{H}_{imp}$$

$$+\sum_i I(\mathbf{r}-\mathbf{r}_i)\psi^\dagger(\mathbf{r})\sigma(g-1)\mathbf{J}_i\psi(\mathbf{r}) + \frac{(rot\mathbf{A}(\mathbf{r}))^2}{8\pi}\} + \sum_i[-\mathbf{B}(\mathbf{r}_i)g_e\mu_B\mathbf{J}_i + \hat{H}_{CF}(\mathbf{J}_i)]. \quad (1)$$

Here, $\epsilon(\hat{\mathbf{p}}-\frac{e}{c}\mathbf{A})$, $\Delta(\mathbf{r})$, $\mathbf{A}$, $I(\mathbf{r})$, V, σ, $\mathbf{J}_i$ and g are the quasiparticle energy, the superconducting order parameter, the vector potential, the exchange integral, the electron-phonon

coupling constant, Pauli matrices, the total angular moment and the Lande factor respectively. The first three terms in Eq. (1) describe the superconducting mean-field Hamiltonian in the magnetic field $\mathbf{B}(\mathbf{r}) = \mathrm{curl}\mathbf{A}(\mathbf{r})$, while the term $\hat{H}_{imp}$ describes the electron scattering on nonmagnetic impurities. The term proportional to $\sigma(g-1)\mathbf{J}_i$ describes the direct EX interaction between electrons and $LM's$, while $\hat{H}_{CF}(\mathbf{J}_i)$ is responsible for the crystal field effects and magnetic anisotropy. Based on Eq. (1) and by using Eilenberger equations one can find the free-energy functional of the coexistence phase in terms of order parameters – see below.

A. Characteristic parameters of $TmNi_2B_2C$

The critical temperature of the transition into the oscillatory magnetic state $T_M \approx 1.5$ K is small compared to the superconducting critical temperature $T_c \approx 11.5$ K and it is of the order of the exchange energy Θ_{ex} - see below. From $\Theta_{ex} = N(0)h_{ex}^2$ we can estimate the exchange interaction between electrons and LM's, which is characterized by $h_{ex} = I(0)(g-1)nJ$, where $N(0)$ is the electronic density of states at the Fermi level (per LM), n is the concentration of LM's. In absence of data on $N(0)$ and Fermi velocity v_F in $TmNi_2B_2C$ we use the band structure value[13] for $LuNi_2B_2C$, where $N(0) \simeq 2.4$ states/eV$\cdot$ Lu atom and $v_{Fx} = v_{Fy} \simeq (2-3)\times 10^7$ cm/sec. This procedure is justified because the f-levels of Lu and Tm ions are weakly coupled to the conduction electrons. The decrease of T_c in the $(RE)Ni_2B_2C$ family is scaled[16] by de Gennes factor $(g-1)^2J^2$ which allows to estimate $\Theta_{ex} = N(0)h_{ex}^2$ and h_{ex}. Namely, the Abrikosov-Gorkov formula $dT_c/dx \simeq -\pi^2\bar{\Theta}_{ex}/2$ for the decrease of T_c in $Lu_{1-x}Tm_xNi_2B_2C$, i.e. $(\Delta T_c/\Delta x)_{\mathrm{Lu-Tm}} \approx T_c^{\mathrm{Lu}} - T_c^{\mathrm{Tm}} \approx 5$ K$\sim 5\bar{\Theta}_{ex}$ gives $\bar{\Theta}_{ex} \sim 1$ K and $h_{ex} \sim 60$ K.

The long-range part of the EM dipole-dipole interaction between LM's is characterized by $\Theta_{em} = 2\pi n\mu^2$, $\mu = g\mu_B J$. The neutron diffraction measurements[1] in $TmNi_2B_2C$ give $\mu \simeq 5\mu_B$, while from the crystallographic structure follows $n \approx 2\cdot 10^{22}$ cm^{-3}, which gives $\Theta_{em} \approx 2$ K. Note that $\Theta_{em} \sim \Theta_{ex} \sim T_M$. From $\Delta_0 \simeq 1.76\ T_c$ one obtains $\xi_0 \simeq 250$ Å, while from magnetization measurements near H_{c2} and from the slope of H_{c2} near T_c (see Ref. 17) it follows $\kappa = \lambda/\bar{\xi} \simeq 7$ and $\bar{\xi} \simeq 110\ A$, where $\lambda \approx 0.62\lambda_L(\xi_0/l)^{1/2}$ and $\bar{\xi} \approx 0.85(\xi_0 l)^{1/2}$.

This gives the mean-free path $l \approx 50$ Åand the London penetration depth $\lambda_L \approx 500$ Å. One can say that the samples studied by Cho *et al.*[17] were in dirty limit where also holds: $(h_{ex}\tau/\hbar)^2 \ll 1$ and $(Ql)^{-2} \ll 1$. .

B. Free-energy functional of the coexistence phase:

Since $Q \ll k_F$ the problem of interplay between S and M is treated[5,8] using the Eilenberger equations for the normal $g_\omega(\mathbf{v},\mathbf{R})$ and anomalous $f_\omega(\mathbf{v},\mathbf{R})$ electronic Green's function. They describe the motion of electrons in the EX field $\vec{h}_{ex}(\mathbf{R}) = \mathbf{c}h_Q \sin Qz$ ($h_Q = h_{ex}S_Q$, $S_Q = |\langle \mathbf{J} \rangle|/J$) - the EX interaction, and in the dipolar magnetic field $\mathbf{B}(\mathbf{r}) = \mathrm{curl}\mathbf{A}(\mathbf{r})$ - the electromagnetic EM interaction. We present only some necessary results for the free-energy (per LM) $F\{\Delta, S_Q, \mathbf{Q}\} = F_s\{\Delta\} + F_M\{S_Q\} + F_{int}\{\Delta, S_Q\}$, where

$$F_s\{\Delta\} = -\frac{1}{2}N(0)\Delta^2 \ln\frac{e\Delta_0^2}{\Delta^2}. \tag{2}$$

Δ is the S order parameter and Δ_0 is the S order parameter in equilibrium and in absence of magnetism. The magnetic part F_M in the mean-field approach is given by

$$F_M\{S_Q\} = -\sum_Q \{[\Theta_0 + \Theta_{ex}(\tilde{\chi}_e(Q) - 1)][\mid \mathbf{S}_{Q,\perp} \mid^2 + \mid \mathbf{S}_{Q,\parallel} \mid^2]$$

$$+ \Theta_{em} \mid \mathbf{S}_{Q,\parallel} \mid^2 + D(\mid S_{x,Q} \mid^2 + \mid S_{y,Q} \mid^2)\} + F_0\{\mathbf{S}_Q\} \tag{3}$$

Here, $D > 0$ and $\mathbf{S}_{Q,\perp}$, $\mathbf{S}_{Q,||}$ are transverse and longitudinal (w.r.t. $\mathbf{Q}$) components of $\mathbf{S}_Q$ respectively, while $F_0\{S_Q\}$ is the isotropic part (entropy term) of the functional for isolated ions. $\Theta_0 = \Theta_{ex} + \Theta_{em}/3 + \Theta'_{ex} + \Theta'_{em}$ characterizes the contribution of all mechanisms (long $(\Theta_{ex}, \Theta_{em})$ - and short $(\Theta'_{ex}, \Theta'_{em})$-range parts of the exchange and dipole energies) to the ground-state energy[5,8]. We assume that in the normal state the dipole-dipole interaction of LM moments leads to ferromagnetic ordering, but exchange interaction may result in ferromagnetic or oscillatory ordering depending on Fermi surface structure. The electronic susceptibility $\chi_e(Q) \equiv N(0)\tilde{\chi}_e(Q)$ is still unknown, although it can be calculated by knowing the band structure of $TmNi_2B_2C$. Two scenarios for $\chi_e(Q)$ will be presented below. The interaction part, $F_{int}\{\Delta, S_Q\} = F_{int}^{ex} + F_{int}^{em}$, of the free-energy contains the EX and EM contributions respectively.

For samples $TmNi_2B_2C$ studied by Cho *et al.*[17] the *dirty limit* ($Q \ll l \ll \xi_0$) is realized, as well as $lh_{ex}S_Q/v_F)^2, (elA_Q/c)^2 \ll 1$, which allows us to find F_{int}

$$F_{int}\{\Delta, S_Q\} \equiv \frac{\pi N(0)\Delta}{2\tau_m} = \frac{\pi^2\Delta}{2}\sum_Q\{\frac{\Theta_{ex}}{v_F Q} \mid \mathbf{S}_Q \mid^2 + \frac{3\Theta_m}{2v_F\lambda_L^2 Q^3} \mid \mathbf{S}_{Q,\perp} \mid^2\}. \quad (4)$$

The first term on the right hand side of Eq. (4) describes F_{int}^{ex} and the second one F_{int}^{em} respectively. Eq. (5) is derived by assuming that: (a) $\tau_m\Delta > 1$ what is indeed fulfilled in $TmNi_2B_2C$, where $\tau_m\Delta \approx 6-7$; (b) the Fermi surface is isotropic - fulfilled also in $TmNi_2B_2C$. Note, the expression for $F_{int}\{\Delta, S_Q\}$ in the clean limit can be found in Ref. 5.

Already on this level we can estimate the relative contribution of the EX and EM terms in the interaction of superconducting and magnetic subsystems in $TmNi_2B_2C$. For parameters extracted from experiments - see A, one gets $r \equiv (F_{int}^{em}/F_{int}^{ex}) \simeq \Theta_{em}/\Theta_{ex}(\lambda_L Q)^2 \approx 10^{-4}$. This important result means that in $TmNi_2B_2C$ the EM interaction makes a *negligible* contribution to F_{int} and as a consequence the competition of S and the M order in $TmNi_2B_2C$ is exclusively due to the *EX interaction* . The similar situation is realized in the whole family of (RE)Ni_2B_2C compounds where $r < 10^{-3}$. Moreover, when $TmNi_2B_2C$ is placed in external magnetic field the EX interaction plays a decisive role- see below. This means that the approach which is based on the EM interaction only (see Ref. 18 and references therein) is inadequate in explaining properties of the (RE)Ni_2B_2C family. However, the EM interaction, although much less detrimental for superconductivity than the EX one, makes the magnetic structure transverse, i.e. $\mathbf{S}\cdot\mathbf{Q} = \mathbf{0}$ due to the $\Theta_{em} \mid \mathbf{S}_{Q,\parallel} \mid^2$ term in Eq. (3).

C. Origin of the oscillatory magnetic order

The magnetic free-energy in the normal state $F_M\{S_Q\}$ depends on the electronic susceptibility $\tilde{\chi}_e(Q) = \chi_e(Q)/g_e^2\mu_B^2 N(0)$ and contains magnetic anisotropy ($D > 0$) and the single ion term $F_0\{S_Q\}$ - see Eq. (3). These quantities determine magnetic structure in $TmNi_2B_2C$ in absence of superconductivity. At present both are unknown and therefore in what follows we analyze two possible scenarios for the origin of the oscillatory magnetic structure, which depends on the form of $\chi_e(Q)$ in $TmNi_2B_2C$:

1. Ferromagnetic (F) scenario In this scenario $\chi_e(Q)$ reaches maximum at $Q = 0$, i.e.

the *ferromagnetic* order would be realized in the normal conduction state of $TmNi_2B_2C$ below some temperature - see Fig. 1a. However, in the S state it is transformed into an oscillating magnetic structure with the wave vector $Q \ll k_F$ - see below, and because $a^2Q^2 \ll 1$ (magnetic length $a < k_F^{-1}$) one has $\chi_e(Q) \approx N(0)(1 - a^2Q^2)$. Replacing this $\chi_e(Q)$ in Eq.(3) and by minimizing the free-energy F with respect to Q one gets the sinusoidal magnetic structure at T very neat T_M (when $h_{ex}S_Q \ll \Delta$ and higher order terms in $\mid \mathbf{S}_Q \mid^2$ are negligible) with $Q_M = (\pi\Theta_{ex}/4\Theta_0 a^2\xi_0)^{1/3}$. In the presence of the magnetic anisotropy and by lowering temperature $\mid \mathbf{S}_Q \mid$ grows and higher order terms in $\mid \mathbf{S}_Q \mid^2$ (described by $F_0\{\mathbf{S}_Q^2\}$ in Eq. (3)) become important giving rise to higher harmonics $3Q, 5Q$, etc.. As a result the striped transverse one-dimensional domain structure ($\mathbf{S}_Q \cdot \mathbf{Q} = 0$, $\mathbf{S}_Q \parallel z$-axis) is formed[5],[8] with the magnetic energy (per LM)

$$F_M = F_0\{S_Q^2\} - \Theta_{ex}S_Q^2 + \eta(S_Q^2, T)\frac{Q}{\pi}, \tag{5}$$

where η is the domain wall energy given by $\eta = k_F^{-1}\Theta_w S_Q^2$. Here, $\Theta_w \approx 0.6(\Theta_0 D)^{1/2}$ for $D < \Theta_0$ but $(D/\Theta_0)^{3/4} > 0.25(k_F\xi_0)^{-1/2}$, while $\Theta_w \approx 0.3\Theta_0$ for $D > \Theta$. This phase is in further called the DS-phase. In the DS-phase and at $T \ll T_M$ the wave vector of the structure is given by $Q_{DS} \approx 2(\Theta_{ex}/\Theta_w k_F\xi_0)^{1/2}$. Since $Q^{\exp} = 0.241$ Å^{-1} and by knowing k_F - for instance $k_F \sim 1$ Å^{-1}, one obtains reasonable value for $\Theta_w \sim (0.1 - 0.2)$ K. These results mean that in the F-scenario the transformation from sinusoidal to the domain-like structure takes place around T_M, with small changes from Q_M to Q_{DS}, where superconductivity and the domain-like magnetic structure (the DS phase) coexist. Moreover, for the given set of parameters in $TmNi_2B_2C$ - see A, one gets that at $T = 0$ one has $F_{DS} - F_M \approx -0.3N(0)\Delta^2/2$, where F_{DS} is the free-energy of the DS-phase. This means that the F-scenario for the origin of the oscillatory magnetic order in $TmNi_2B_2C$ predicts that superconductivity *coexists* with the domain-like magnetic order up to $T = 0$. We pay attention that the latter result is independent of the scenario (F or O - see below) and it is in accordance with the experimental finding[1] in $TmNi_2B_2C$, where S and the oscillatory magnetic order coexist up to $T = 0$.

II. O-scenario - In this scenario it is assumed that the *oscillatory* magnetic order with the

wave vector Q would be realized in absence of superconductivity, i.e. $\chi_e(Q)$ is peaked at Q_0 - see Fig. 1b. At lower temperatures the magnetic anisotropy and the single ion term $F_0\{S_Q^2\}$ transform the structure into a domain-like one. In this case $F_{int}\{\Delta, \mathbf{S}_Q, \mathbf{Q}\}$ is also given by Eq. (4), where the wave-vector Q should be considered fixed (by experiment). We point out that the domain-like magnetic structure in the O-scenario is a property of the normal state and not of the superconducting one - see *Fig.*1*b*. Note, the ratio $r \ll 1$, i.e. it is small in both scenarios. Because in the O-scenario one has also $F_{DS} - F_M \approx -0.3N(0)\Delta^2/2$ then the domain-like magnetic structure and superconductivity coexist also up to $T = 0$.

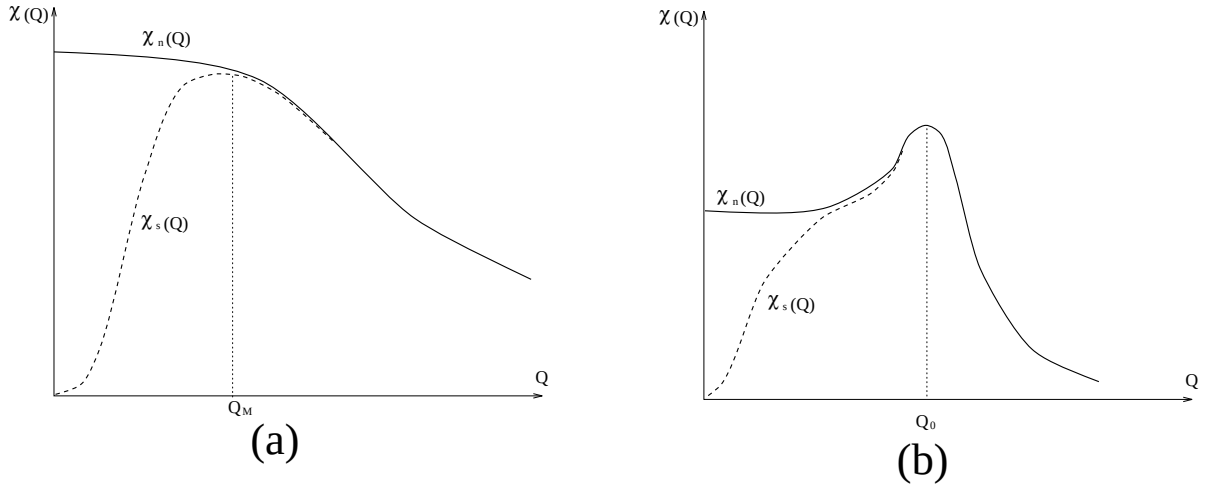

FIG. 1. Schematic shape of the spin susceptibility of conduction electrons in the normal - $\chi_n(Q)$ and superconducting - $\chi_s(Q)$ state in the case of: (a) the F-scenario, where the peak in $\chi_s(Q)$ is due to superconductivity; (b) the O-scenario, where the peak in $\chi_s(Q)$ is independent on superconductivity.

Note, recent neutron scattering measurements show[2] clearly the presence of the third harmonic ($3Q$), at $T < 1$ K, with the intensity $I_{3Q} \approx 0.03 I_Q$ what tells us that the magnetic structure in $TmNi_2B_2C$ is domain-like. The smallness of I_{3Q} can be due to the presence of defects in the sample which are always detrimental for a domain structure[5,19]. The F- and O-scenario can not be resolved by this type of measurement. For that we need to study the system in magnetic field.

D. Gapless superconductivity

In clean superconductors the oscillatory magnetic order can give rise to the gapless quasiparticle spectrum[5,8,9] if $h_{ex} > \Delta$, what is just the case in $TmNi_2B_2C$ where $h_{ex} > 60$ K and $\Delta < 20$. The gapless region on the Fermi surface given by the condition $\mathbf{Q} \cdot \mathbf{v}_F = 0$. In the DS-phase at temperatures where $h_{ex} S_Q > \Delta$ the density of states is given by

$$N_s(E) = N(0) \frac{h_{ex} S_Q}{\Delta \cdot v_F Q} E \ln \frac{4\Delta}{\pi E}, \qquad E \ll \Delta. \tag{6}$$

By measuring tunneling conductance, where $\sigma(V) \sim N_s(V)$, one could test this prediction which is a consequence of the EX interaction.

E. Effects in magnetic field

Measurements in magnetic field can discern between F- and DS-scenarios. In the F-scenario the critical field H_c^{FS} for the first order F-DS transition (at $T \ll T_M$) is obtained from the condition $-HM(0) = -N(0)\Delta_0^2/2$, i.e. $H_c^{FS} \approx 200$ G for $M(0) = 10^3$ G ($M(0) = n\mu$ is the saturation magnetization). Note, in getting H_c^{FS} we have assumed that the field is oriented along the c-easy axis while if it is along the hard axis it could be much higher, i.e. $H_{a-b}^{FS} \gg H_c^{FS}$. On the first sight such a small value of H_c^{FS}contradicts reports on the critical field[16] in $TmNi_2B_2C$, where rather high critical field $H_{c2} \sim 1$ T is found near T_M. Concerning this point one should stress the measurements[16] have been done: (1) on the polycrystals; (2) at fixed magnetic field H by lowering temperature. Because of possibility $H_{a-b}^{FS} \gg H_c^{FS}$ the measurements on single crystals are desired, and because the transition at H_c^{FS} is of the first order, with a possibility for huge hysteresis, one should perform measurements at fixed T in increasing and decreasing field. Note, such a huge hysteresis is not expected in the O-scenario, where the critical magnetic field is determined by superconducting properties mainly and must be much larger than H_c^{FS}.

In conclusion, we have found that the oscillatory (domain-like) magnetic order and superconductivity coexist in $TmNi_2B_2C$ up to $T = 0$ and that their competition is due to the exchange interaction between conduction electrons and localized moments (LM's), while the electromagnetic interaction is negligible, eventually helping in making the magnetic structure

transverse. The type and the origin of the magnetic structure in absence of superconductivity can be resolved by measuring critical magnetic fields, where a huge hysteresis could favor the F-scenario, i.e. the ferromagnetic order in absence of superconductivity. It is predicted the gapless superconductivity in clean $TmNi_2B_2C$ with line of zeros on the Fermi surface.

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